

Magnetic domain fluctuations in an antiferromagnetic film observed with coherent resonant soft x-ray scattering

S. Konings,¹ C. Schüßler-Langeheine,² H. Ott,² E. Weschke,³ E. Schierle,³ and J. B. Goedkoop¹

¹*Van der Waals-Zeeman Institute, University of Amsterdam, 1018 XE Amsterdam, The Netherlands*

²*II. Physikalisches Institut, Universität zu Köln, Zùlpicher Str. 77, 50937 Köln, Germany*

³*Institut für Experimentalphysik, Freie Universität Berlin, 14195 Berlin, Germany*

(Dated: May 25, 2008)

We report the direct observation of slow fluctuations of helical antiferromagnetic domains in an ultra-thin holmium film using coherent resonant magnetic x-ray scattering. We observe a gradual increase of the fluctuations in the speckle pattern with increasing temperature, while at the same time a static contribution to the speckle pattern remains. This finding indicates the coexistence of static and fluctuating domains in the film. We ascribe this non-ergodic behavior to thickness variations caused by steps at the interfaces.

PACS numbers: 75.10.Nr, 75.70.Rf, 78.70.Ck

Slow dynamics of magnetic domains on time scales of nanoseconds and longer are of high practical importance. Domain wall dynamics play a crucial role in magnetization reversal processes; thermally activated domain wall motions determine the lifetime of magnetically stored information. Slow dynamics on nanometer length scales is best probed by x-ray photon correlation spectroscopy (XPCS) [1, 2, 3, 4, 5, 6, 7, 8, 9, 10] using the coherent diffraction or *speckle* pattern, which is generated when a coherent light beam scatters from a disordered structure. Any fluctuations in the disorder lead to a change in the speckle pattern; the dynamics in the sample can be obtained by measuring the time averaged intensity correlation function (ICF) [11, 12, 13] of the speckle intensities on time scales ranging from 50 ns [6] to hours [4, 10]. Importantly, with PCS one can obtain directly the fluctuating and static parts of the sample [12, 13], which makes PCS highly attractive for the investigation of systems where pinning or jamming effects occur [4].

In order to study magnetism the speckle experiment has to be sensitive to spin degrees of freedom. X-ray scattering in the conventional x-ray range, even at electronic resonances, has a low magnetic scattering cross section [14]. The only exception are $5f$ systems where a magnetic phase transition in UAs has indeed been observed by the loss of speckle contrast [15]. But actinide systems are only of limited practical relevance, whereas in most interesting magnetic systems the magnetism is carried by $3d$ or $4f$ electrons. $4f$ magnetism can be probed at the $2p \rightarrow 5d$ resonances in the conventional x-ray range, but such experiments usually require polarization analysis of the scattered photons, which is hard to combine with the high spatial resolution required to resolve the speckle pattern. The scattering cross section for $3d$ magnetism on and off the $1s \rightarrow 4p$ resonance is very low and only few magnetic scattering studies of $3d$ transition-metal systems in the conventional x-ray range exist.

In some cases one may probe magnetism indirectly, via

its coupling to structural degrees of freedom [10]. But while a coupling of spin order to a CDW or to charge order is found for many systems, both orders will generally form on different temperature scales. Examples are layered nickelates [16] or cobaltates [17], for which the temperature scales for charge and for spin order are clearly different. An extreme case is $\text{La}_{1.5}\text{Sr}_{0.5}\text{CoO}_4$, where charge order sets in below 750 K, while static spin order is not observed above 35 K [17]. It is therefore not to be expected that the charge dynamics are generally representative for the dynamics of magnetic order. This means that the indirect approach cannot be generally applied and one needs to probe the magnetic signal directly.

This is possible in soft x-ray range where resonant scattering provide a high magnetic contrast. PCS at the $\text{Co } 2p \rightarrow 3d$ resonance was used to study the influence of disorder on the static domain pattern of Co/Pt multilayers for different magnetic fields [18]. In this Letter we show that using soft x-ray PCS it is actually possible to directly probe *fluctuating* magnetic domains near a second-order phase transition and to address the question, how this transition is affected by static disorder in the system.

We studied an 11 monolayers (ML) thin epitaxially grown Ho-metal film sandwiched between Y-metal layers [19, 20]. Holmium metal displays a helical magnetic phase (sketched in Fig. 1(a)) over a wide temperature range leading to superstructure peaks in the magnetic diffraction signal separated by a wavevector $(0, 0, \pm\epsilon)$ from the structural peaks [21]. The ordering temperature T_N depends on the film thickness such that films below 10 ML, which is about one helix period length in bulk Ho, do not show any helical order [22]. Our film is hence near the stability limit for helical order and thus close to two-dimensionality. On the other hand such a film should be very susceptible to slight thickness variations because T_N is a steep function of the thickness. From reflectivity measurements we indeed find a rough-

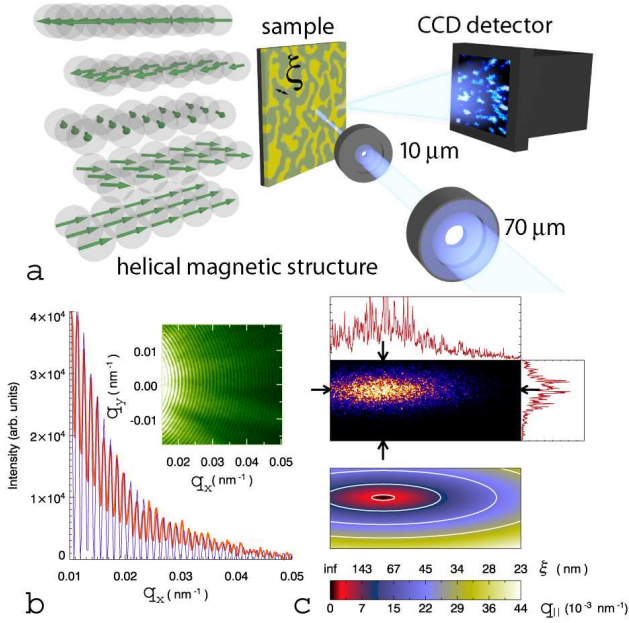


FIG. 1: (a) Sketch of the scattering experiment and the helical antiferromagnetic structure. Two pinholes select the spatially coherent part of the undulator radiation. The correlation length ξ in the sample is indicated with a black line and is in reality ~ 100 times smaller than the beam diameter. (b) The diffraction pattern of the $10\ \mu\text{m}$ pinhole. q denotes the momentum transfer. A line cut through the center is shown in orange, together with a least squares fit of an Airy pattern in blue. (c) A typical speckle pattern of the magnetic satellite peak at 30 K. The black arrows indicate the positions of the two intensity line traces plotted in red. The corresponding color-coded scale of the correlation length ξ or the in-plane momentum transfer $q_{||}$ is also displayed.

ness of the Ho/Y interfaces of the order of 2 ML.

The experiments were carried out at the BESSY U49/2-PGM1 beam line and at the UE46 beam line operated by the Hahn-Meitner-Institute at BESSY using the soft x-ray diffractometer built at the FU Berlin. In order to observe the magnetic signal from the film, we used the strong magnetic contrast that is found at the $3d \rightarrow 4f$ (M_5) excitation in the soft x-ray range at a photon energy around 1344 eV corresponding to a photon wavelength $\lambda=9.2\ \text{\AA}$ [19, 23]. We intercepted the first-order (00ϵ) satellite with a direct-exposure soft x-ray CCD camera as displayed in Fig. 1(a). With incoherent light, we observe a smooth diffraction peak. As is shown in Fig. 2 (diamond symbols), the scattered intensity in this peak decreases when the nominal transition temperature $T_N=76\ \text{K}$ is approached, but does not vanish up to $\approx 90\ \text{K}$ [24]. The peak profile is well described by a single Lorentzian, with a half-width $W_{q_{||}}$ that equals the inverse of the in-plane correlation length ξ of the magnetically ordered regions. As shown by the circular symbols in Fig. 2, we find ξ to decrease from

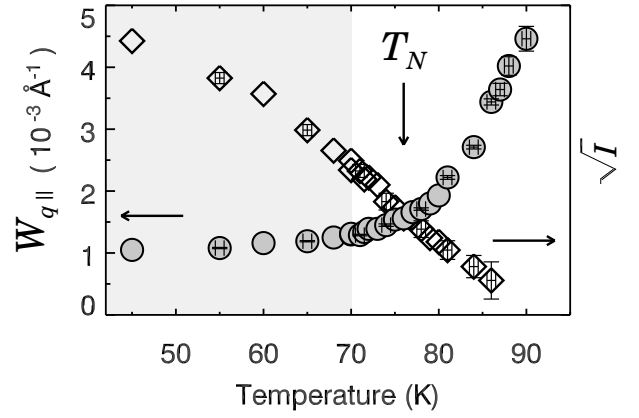


FIG. 2: Half-width of the magnetic satellite peak $W_{q_{||}}$ (circles and left axis) and square root of the integrated satellite intensity (diamonds and right axis) as measured with a non-coherent beam. The grey area indicates the temperature range that could be studied with XPCS.

90 nm at 45 K to 30 nm at 90 K. The loss of coherence already sets in around 50 K and $W_{q_{||}}$ keeps on changing over an unusually wide temperature interval of more than 40 K. This may either be an intrinsic effect caused by the proximity of the film to two-dimensionality or an effect of (static) disorder induced by the interface roughness. In order to disentangle the roles of static and dynamic effects at this second order phase transition, we selected the transversely-coherent fraction of the BESSY II undulator radiation using a $10\ \mu\text{m}$ pinhole in front of the sample. This causes the smooth magnetic diffraction peak to break up into myriad speckles [Fig. 1(c)], which form the diffraction pattern of the magnetic domain structure of the particular illuminated spot. Speckle fluctuations at different distances from the peak center are related to real-space fluctuations on different length scales [Fig. 1(c)]. We obtain the most intense signal from magnetic disorder on length scales of more than 100 nm, which we assign to helicity or phase domains.

We followed the time evolution of the speckle pattern by recording movies over a period of several hours with exposure times of 4 or 10 seconds. Snapshots from these movies for 52 K and 70 K are presented in the small frames of Fig. 3. The complete movies for various temperatures are available online [25]. At 52 K the speckle pattern is static on a time scale of one hour. At intermediate temperatures the speckle pattern starts to change with time and already at 70 K the movement of the speckles is very vivid. In order to quantify how much of the speckle pattern is moving, we took the time average of all the frames in the movie, shown in the large panels of Fig. 3. At 52 K the average pattern is equal to that of a single frame. Closer to the phase transition subsequent speckle patterns differ strongly, and the time-averaged speckle pattern is much smoother than the individual frames thus showing that domain-wall fluctuations have

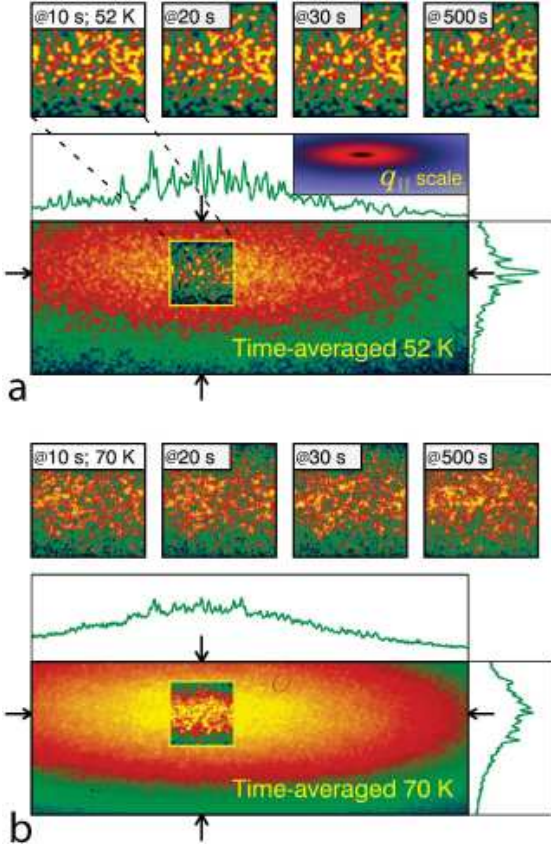


FIG. 3: Time-averaged intensity distribution of the magnetic satellite peak (*large panels*) at 52 K (a) and 70 K (b). A logarithmic color scale is used to better observe the speckle at higher $q_{||}$. The snapshots (*small panels*) are single frames with an exposure time of 10 s taken at the indicated time. For the snapshots a linear color scale is used. The $q_{||}$ -scale corresponds to the ones in the large panels and is color coded as in Fig. 1(c).

started. But even the time averages of films at higher temperatures over hours show some graininess due to the existence of static speckles connected to non-fluctuating parts of the domain pattern. These static speckles are found on all length scales, i.e. at all distances from the peak center, which is very obvious in the line cuts in Fig. 3(b). Our finding thus implies that some regions of the sample are fluctuating, while others remain fixed over the measurement period: the system behaves non-ergodic.

The most likely cause for the observed coexistence of static and fluctuating domains are steps at the Ho/Y interfaces, which cause variations in the film thickness. As noted above, the ordering temperature T_N of Ho films is critically dependent on the film thickness in the range of 10 to 12 monolayers [22]. This leads to a picture in which at low temperatures the magnetization has settled down in an irregular static domain structure. As the temperature increases, the thinnest regions approach their local

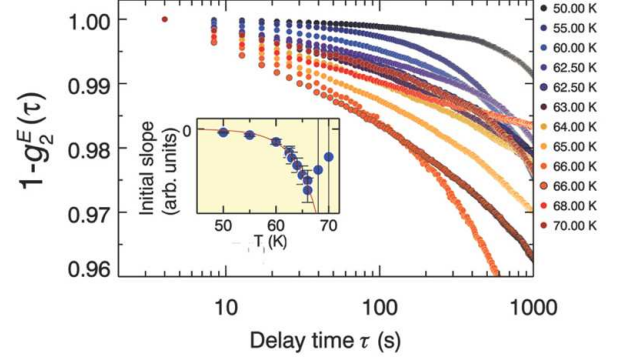


FIG. 4: Normalized experimental results for $1 - g_2^E(\tau)$ at the indicated temperature. The inset shows the initial slope for each temperature, with an exponential fit (*red line*).

Néel temperature and start to fluctuate. At higher temperatures, gradually the thicker regions join in, explaining the observed reduction of the magnetic correlation length and the increasing fluctuation rates in the speckle movies.

The increase of the fluctuation rates with temperature are also reflected in the time-averaged intensity correlation function (ICF). We performed the ensemble averaging required for non-ergodic systems [12, 13] by averaging over all the pixels with a distance from the peak center of less than 0.004 nm^{-1} corresponding to correlation lengths of 250 nm and larger. The normalized ensemble averaged ICF is defined as $g_2^E(\tau) = \langle I(t)I(t+\tau) \rangle_E / \langle I(t) \rangle_E^2$, where τ gives the delay time between two data samples and the brackets $\langle \rangle_E$ indicate time and ensemble averaging. In Fig. 4 we show the results for $1 - g_2^E(\tau)$ for the different temperatures, with the curves normalized to the first data point. For short correlation times the signal changes faster and faster with τ upon heating (see inset). This reflects an increase of the domain wall dynamics that speeds up with increasing temperature. The last two points in the inset indicate that for temperatures above 66 K the scattered intensity becomes too low for a reliable analysis. [26, 27]

In conclusion, we show that already with the limited coherent flux available at present 3rd-generation light sources, x-ray photon correlation spectroscopy at soft x-ray resonances provides unique information on the influence of disorder on magnetic phase transitions. The observed non-ergodic behavior indicates that the phase transition in an ultrathin Holmium film is dominated by pinning of the magnetic fluctuations in the potential landscape formed by local thickness variations. This image also provides a natural explanation for the smearing out of the magnetic phase transition over a wide temperature range found for this sample.

Presently, soft x-ray resonant PCS experiments are limited by the available coherent flux. With the new x-ray free-electron laser sources, which deliver a fully co-

herent photon beam, the extension of the experiment to weaker signals and higher temperatures is straightforward [28]. Resonant soft x-ray scattering is not only sensitive to $4f$ and $3d$ magnetism, but also to charge and orbital order with an equally high sensitivity and scattering cross section. Soft x-ray PCS hence provides the ideal tool for the study of thermal evolution of disorder, fluctuations and pinning effects in a wide variety of solid state systems with magnetic or electronic correlations such as nano-crystalline and cluster systems, charge- and spin density waves, stripe- and orbital order in correlated electron systems.

The authors thank H. Zabel for providing the sample, the BESSY and HMI staff of U49/2-PGM1 and UE46-PGM, notably D. Schmitz for expert assistance during the measurements. We thank G. Wegdam and D. Bonn for discussions on PCS. The authors gratefully acknowledge financial support by the Stichting voor Fundamenteel Onderzoek der Materie (FOM), the Deutsche Forschungsgemeinschaft (DFG) through SFB 608, the BMBF through project 05 ES3XBA/5, and BESSY as a partner of the EU's I3 IA-SFS project (BESSY-ID.06.2.198; RII 3CT-2004-506008).

-
- [1] S. B. Dierker, R. Pindak, R. M. Fleming, I. K. Robinson, and L. Berman, Phys. Rev. Lett. **75**, 449 (1995).
 - [2] T. Thurn-Albrecht, W. Steffen, A. Patkowski, G. Meier, E. W. Fischer, G. Grübel, and D. L. Abernathy, Phys. Rev. Lett. **77**, 5437 (1996).
 - [3] S. G. J. Mochrie, A. M. Mayes, A. R. Sandy, M. Sutton, S. Brauer, G. B. Stephenson, D. L. Abernathy, and G. Grübel, Phys. Rev. Lett. **78**, 1275 (1997).
 - [4] A. Malik, A. R. Sandy, L. B. Lurio, G. B. Stephenson, S. G. J. Mochrie, I. McNulty, and M. Sutton, Phys. Rev. Lett. **81**, 5832 (1998).
 - [5] T. Seydel, A. Madsen, M. Tolan, G. Grübel, and W. Press, Phys. Rev. B **63** 073409 (2001).
 - [6] I. Sikharulidze, I. P. Dolbnya, A. Fera, A. Madsen, B. I. Ostrovskii, and W. H. de Jeu, Phys. Rev. Lett. **88** 115503 (2002).
 - [7] I. Sikharulidze, B. Farago, I. P. Dolbnya, A. Madsen, and W. H. de Jeu, Phys. Rev. Lett. **91**, 165504 (2003).
 - [8] A. Madsen, T. Seydel, M. Sprung, C. Gutt, M. Tolan, and G. Grübel, Phys. Rev. Lett. **92** 096104(2004).
 - [9] A. Madsen, T. Seydel, M. Tolan, and G. Grübel, J. Synchr. Rad. **12**, 786 (2005).
 - [10] O. G. Shpyrko, E. D. Isaacs, J. M. Logan, Y. Feng, G. Aepli, R. Jaramillo, H. C. Kim, T. F. Rosenbaum, P. Zschack, M. Sprung, S. Narayanan, and A. R. Sandy, Nature **447**, 68 (2007).
 - [11] B. J. Berne and R. Pecora, *Dynamic light scattering* (Dover publications Inc., Mineola, New York, 2000).
 - [12] P. N. Pusey and W. van Megen, Physica A **157**, 705 (1989).
 - [13] M. Kroon, G. H. Wegdam, and R. Sprik, Phys. Rev. E **54**, 6541 (1996).
 - [14] C. Vettier, Journ. Electron. Spectrosc. Relat. Phenom **117-118**, 113 (2001).
 - [15] F. Yakhou, A. Letoublon, F. Livet, M. de Boissieu, and F. Bley, J. Magn. Magn. Mater. **233**, 119 (2001).
 - [16] H. Yoshizawa, T. Kakeshita, R. Kajimoto, T. Tanabe, T. Katsufuji, and Y. Tokura, Phys. Rev. B **61**, R854 (2000).
 - [17] I. A. Zaliznyak, J. P. Hill, J. M. Tranquada, R. Erwin, and Y. Moritomo, Phys. Rev. Lett. **85**, 4353 (2000).
 - [18] M. S. Pierce, C. R. Buechler, L. B. Sorensen, J. J. Turner, S. D. Kevan, E. A. Jagla, J. M. Deutsch, T. Mai, O. Narayan, J. E. Davies, K. Liu, J. H. Dunn, K. M. Chesnel, J. B. Kortright, O. Hellwig, and E. E. Fullerton, Phys. Rev. Lett. **94**, 017202 (2005).
 - [19] H. Ott, C. Schüßler-Langeheine, E. Schierle, A. Y. Grigoriev, V. Leiner, H. Zabel, G. Kaindl, and E. Weschke, Phys. Rev. B **74**, 094412 (2006).
 - [20] V. Leiner, D. Laberge, R. Siebrecht, C. Sutter, and H. Zabel, Physica B **283**, 167 (2000).
 - [21] G. Helgesen, J. P. Hill, T. R. Thurston, D. Gibbs, J. Kwo, and M. Hong, Phys. Rev. B **50**, 2990 (1994).
 - [22] E. Weschke, H. Ott, E. Schierle, C. Schüßler-Langeheine, D. V. Vyalikh, G. Kaindl, V. Leiner, M. Ay, T. Schmitte, H. Zabel, and P. J. Jensen, Phys. Rev. Lett. **93**, 157204 (2004).
 - [23] S. Konings, PhD thesis, University of Amsterdam (2007).
 - [24] H. Ott, PhD thesis, Freie Universität Berlin (2004).
 - [25] <http://www.science.uva.nl/research/cmp/konings/speckle>.
 - [26] D. Lumma, L. B. Lurio, S. G. J. Mochrie, and M. Sutton, Rev. Sci. Instr. **71**, 3274 (2000).
 - [27] P. Falus, M. A. Borthwick, and S. G. J. Mochrie, Review of Scientific Instruments **75**, 4383 (2004).
 - [28] G. Grübel, G. B. Stephenson, C. Gutt, H. Sinn, and T. Tschentscher, Nucl. Instr. Meth. B **262**, 357 (2007).